

Electronic state of a doped Mott-Hubbard insulator at finite temperatures studied using the dynamical mean-field theory

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Abstract

We study the electronic state of the doped Mott-Hubbard insulator within Dynamical Mean Field Theory. The evolution of the finite temperature spectral functions as a function of doping show large redistributions of spectral weight in both antiferromagnetic and paramagnetic phases. In particular, a metallic antiferromagnetic state is obtained with a low frequency Slater-splitting quasiparticle peak coexisting with Hubbard bands. In the high temperature paramagnetic metallic phase, upon reducing doping, the system has a crossover through a “bad metal” state characterized by an anomalous shift of the quasiparticle peak away from the Fermi energy. We find that the *charge* compressibility of the antiferromagnetic metal is dramatically enhanced upon approaching the second order Néel line.

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The understanding of metal-insulator transitions is a central problem of condensed matter physics. One of the most intriguing is the Mott transition, where the itinerant electrons of a metal localize due to strong correlation effects stemming from Coulomb repulsion [1]. In a Mott state, though the electrons become localized, their spin (and possibly orbital) degrees of freedom remain unquenched. This leads to a large entropy and consequently, Mott insulators tend to order magnetically (and/or orbitally) at sufficiently low temperatures [2]. Examples of strongly correlated systems where the Mott phenomenon occur include materials with partially filled d and f orbitals. The Mott transition can be driven by pressure, temperature or doping and exotic physics such as high temperature superconductivity, colossal magnetoresistance and heavy fermion states are usually found in the vicinity of Mott insulating states [2].

A minimal model that captures the physics of the Mott transition is the Hubbard model. The development of the Dynamical Mean Field Theory (DMFT)[3] enabled progress in the theoretical understanding of the Mott-Hubbard transition in the limit of large lattice coordination. Moreover, the combination of DMFT with ab-initio methods is a promising new tool to study strongly correlated real materials [4] which are often described by complex multi-band hamiltonians. In the light of these developments, a clear and reliable solution of basic models like the Hubbard model is an indispensable stepping stone. Much of the work has been centered on the study of the *frustrated* Hubbard model which has no anti-ferromagnetic (AF) order and shows a first order Mott transition line that ends in a finite temperature second order critical point [3, 5, 6]. The original Hubbard model, however, has a Mott insulating phase at low temperature with Néel order. This AF order can be destroyed by increasing temperature, leading to a paramagnetic (PM) Mott insulator; or by doping, leading to a strongly correlated PM metal [3]. The dramatic changes of the electronic state of the system as it is doped away from the Mott state to the correlated metallic state are of great current interest. For example, cuprates first go through an intriguing pseudogap state [7] followed by a strange metallic one with anomalous properties, to end in a strongly correlated Fermi liquid [7, 8, 9]. Other compounds like the titanates LaTiO_3 and CaTiO_3 and the vanadate V_2O_3 , also exhibit anomalous behaviors with doping, such as a divergent effective mass and large transfers of spectral weight [2].

Here, we focus on the finite temperature doping driven metal-insulator transition in the original Hubbard model, i.e. *without frustration*. We present a detailed solution of the

model with particular emphasis on the systematic changes of the density of states (DOS). A highly correlated AF metal phase is found and contrary to naive expectations, the onset of magnetism has a dramatic effect on the electronic compressibility. We find that the high temperature PM metallic phase also shows novel behavior at very low doping. These properties are clearly relevant for interpretations of recent experiments in photoemission where the dependence of the doping on chemical potential was precisely measured [9], and also for STM spectroscopies and ultra-sound velocity attenuation amongst others.

The Hubbard model reads,

$$H = t \sum_{\langle ij \rangle \sigma} [c_{i\sigma}^+ c_{j\sigma} + h.c.] - \mu \sum_{i\sigma} n_{i\sigma} + U \sum_i (n_{i\uparrow} - \frac{1}{2})(n_{i\downarrow} - \frac{1}{2}) \quad (1)$$

where the hopping t is between nearest neighbors, $c_{i\sigma}^+$ creates a particle with spin σ at site i , U is the Coulomb repulsion and μ is the chemical potential. In the limit of large lattice coordination, the above model can be exactly mapped onto a single impurity Anderson model with a supplementary self-consistency condition for the hybridization function [3]. For simplicity, we consider a Bethe lattice which is bipartite and has a semicircular density of states, and therefore, well adapted to the study of commensurate magnetism. The results are expected to be qualitatively similar on a hypercubic lattice.

The magnetic phase diagram of the model on a hypercubic lattice as a function of doping was obtained in [10] by computing the spin susceptibility in the PM phase. In that work, it was found that the AF instability is towards a commensurate state for most of the parameter space. The instability towards incommensurate states was found at very low temperatures and for a small range of doping. In contrast to Ref.[10], in this Letter, we consider Néel AF order and we explicitly solve for the DMFT equations in the broken symmetry phase. This allows us to directly obtain the spin dependent local Green's function and hence the DOS. Our work does not touch upon the question of phase separation at very low temperatures since we work in temperature ranges and strong coupling where stable commensurate spin order is expected to occur. The DMFT is not the optimal method to study inhomogeneous phases and other exotic orderings. [11]

When AF Néel long range order is allowed on the original lattice, the DMFT self-consistency condition for the associated Anderson impurity model reads [3],

$$\mathcal{G}_{\sigma\sigma}^{-1}(i\omega_n) = i\omega_n + \mu - t^2 G_{-\sigma}(i\omega_n) \quad (2)$$

$\mathcal{G}_{o\sigma}$ and G_σ are the Green's functions of the associated impurity. $t^2 G_{-\sigma}(i\omega_n)$ plays the role of the impurity hybridization $\Delta_\sigma(i\omega_n)$. When the self-consistency condition (2) is fulfilled, the impurity Green's function G_σ coincides with the local Green's function of the lattice [3]. We use the quantum Monte Carlo method of Hirsch and Fye [12] to obtain the exact solution (in the statistical sense) of the model in the strong correlation regime, which is beyond the scope of analytic methods. In order to obtain the DOS, one has to analytically continue the Monte Carlo data to the real axis using a maximum entropy method [13]. Due to the critical fluctuations near the AF/PM boundary, expensive simulations are required to obtain numerically reliable results; we, therefore, focus on a single value of the Coulomb repulsion $U = 3.125$ and the unit of energy is set by the half-bandwidth $D = 2t = 1$. As expected, we find that at half-filling ($\mu = 0$) the system is an AF Mott insulator with a large gap ($\sim D$) below a Néel temperature of $T_N \simeq 0.1$, and a PM Mott insulator above.

We begin with the discussion of the evolution of the DOS as a function doping. We focus on two temperatures which are representative of the two characteristic evolutions of the DOS: $T < T_N$ allows us to trace the evolution of the system from an AF Mott insulator at half-filling to a strongly correlated PM metal as it traverses a second order AF/PM transition line; $T > T_N$ permits us to study the *crossover* from the PM Mott insulator to the strongly correlated PM metal through a “bad metal” state.

In Fig.1, we consider $T = 0.0625 < T_N$ and show the DOS for the two spin projections and different values of the chemical potential μ . We consider the case of hole doping, therefore we take $\mu \leq 0$. The top most spectra (E) corresponds to the half-filled AF Mott insulator (AF-I) at $\mu = 0$ and displays up-down symmetry. This state has a rather large staggered moment m (inset) favored by the bipartite nature of the lattice and the large value of U/D . Expectedly, the spectra shows Hubbard bands separated by a large gap of size $\approx 2Um$ (indicated in the figure by double head arrows). The lower edges of the Hubbard bands are strongly enhanced due to the effective doubling of the unit cell in the Néel state. As the inset shows, the chemical potential can be varied within the large Mott gap ($|\mu| \lesssim 0.6$) without changing the filling and the staggered magnetic moment m displaying a plateau. Accordingly, one finds that the DOS merely shows a rigid shift in energy of the spectra E. When μ reaches the Hubbard band edges, carriers are doped to the system and dramatic changes in the spectral functions are observed. They can be more easily understood by following the evolution of the DOS from the high doping PM end, i.e., from the PM metallic

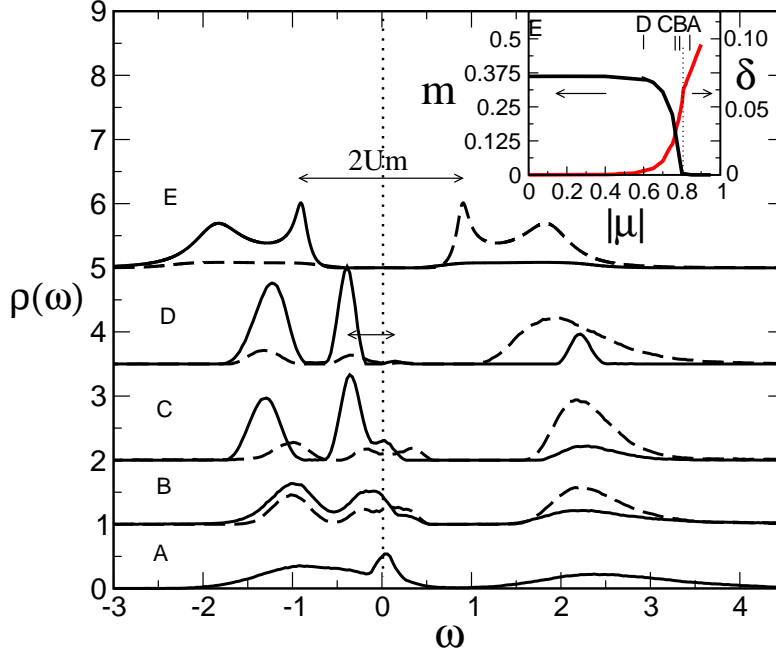


FIG. 1: The local DOS $\rho(\omega)$ for $U = 3.125$ and $T = 0.0625$ for values of $-\mu = 0.84, 0.8, 0.777, 0.6, 0.0$ which correspond to dopings $\delta = 0.076, 0.053, 0.037, 0.003, 0.0$ respectively from bottom to top. Each line type corresponds to a spin projection. The double arrow head line indicates the large Mott gap (E) and its dramatic reduction upon infinitesimal doping (D). The inset shows the staggered magnetization and the doping as a function of $|\mu|$. The letters A to E indicate the values of μ that corresponds to the spectra in the main panel. The dotted line indicates the AF/PM boundary.

spectra A. It is a strongly correlated PM metallic state that shows a narrow quasiparticle peak at the Fermi energy flanked by the lower and upper Hubbard bands. The quasiparticle peak carries a reduced spectral weight and its small width defines the renormalized Fermi energy ϵ_F^* . Due to its proximity to a Mott transition, the effective mass of this state is greatly enhanced compared to the non-interacting value and one has a heavy Fermi liquid (PM-FL) [3, 14].

The spectra B to D show the dramatic changes of the DOS as the system evolves between the two limit cases A and E just described. Reducing δ from the PM metal brings the system back to the AF phase. The localized (Hubbard bands) and itinerant (quasiparticle

peak) aspects of the DOS that coexisted in the PM metallic correlated state survive the AF transition. However, as one enters into the AF state they evolve in a qualitatively different manner: while the incoherent Hubbard bands become polarized and merely modify their relative spectral weight, the coherent quasiparticle peak reveals a Slater like splitting. The latter feature can be understood in two complementary ways: one is to recall that in terms of the associated impurity model, the central peak in the correlated PM metal (spectra A) corresponds to a Kondo resonance. On the AF side, the polarization of the hybridization function $\Delta_\sigma(i\omega_n)$ (cf.(2)) can be interpreted as an effective (frequency dependent) magnetic field at the impurity site. This effective field then induces a Zeeman splitting of the Kondo peak. Another way to understand the behavior of the AF state is to think of the quasiparticle peak as a band narrowing due an effective renormalization of hopping by the interaction, similar to the Brinkman-Rice solution of the Hubbard model [3, 15]. Then, the AF splitting of the quasiparticle band can be simply understood as a Slater-like bandstructure effect due to the doubling of the lattice constant when Néel magnetic order sets in. The splitting of the quasiparticle peak is quite small and the DOS for both up and down spin projections are always *metallic* (AF-M). This observation is qualitatively consistent with the unexpected metallic behavior seen in large regions of the AF phase in the cuprate $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and the insensitivity of the resistivity across the PM/AF transition in clean and very low doped samples ($\sim 1\%$) reported in Ref.[8].

Reducing the doping further, i.e., approaching the $\delta \rightarrow 0$ limit, enhances the asymmetry of the quasiparticle splitted band until the $\omega > 0$ side is nearly suppressed (spectra D). Significantly, the Fermi energy gets within the now clean small gap between the splitted band (due to its weak intensity, the peak in the $\omega > 0$ side can be barely distinguished so is indicated by a double arrow head line in the spectra D) [16]. The system has now become an AF insulator, but qualitatively different from the large gap Mott state E. Interestingly, the staggered moment of this very lightly doped state has almost reached the saturation value, so the small gap value is not given by $2Um$. Finally, when the doping vanishes, the strongly correlated state with low frequency features is no longer sustainable and the spectra undergoes a rapid redistribution of spectral weight to acquire the line shape of E (and rigidly shifted by μ as discussed before).

We now study the *crossover* from the Mott insulator to the strongly correlated metal induced by doping in the higher temperature PM phase. Previous studies of the fully

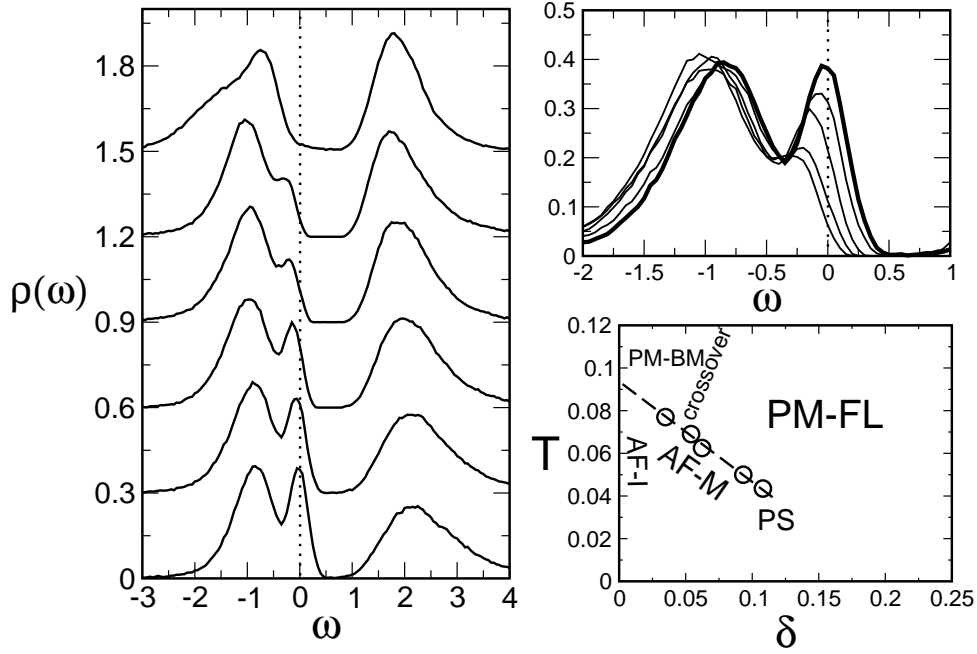


FIG. 2: Left panel: the local DOS $\rho(\omega)$ for $U = 3.125$ and $T = 0.1$. The values of the chemical potential are $-\mu = 0.4, 0.5, 0.55, 0.65, 0.73$ and 0.8 , that correspond to $\delta \approx 0.003, 0.0076, 0.0114, 0.022, 0.038$ and 0.055 respectively (top to bottom). Top right: the low energy part of the same results ($-\mu = 0.4$ is not included and $-\mu = 0.8$ is in thick line). Bottom right: Schematic phase diagram. The circles denote our results for $T_N(\delta)$ and are consistent with those of Ref.10 obtained by computing the susceptibility.

frustrated model, which has no AF order, showed that at low T , the width of the quasiparticle peak is controlled by the doping δ [3, 14, 17, 18]. Since the renormalized Fermi energy $\epsilon_F^*(\delta) \sim \delta D$, it is expected that below the coherence temperature, i.e., when $T < \epsilon_F^*(\delta)$ the system should be a Fermi liquid, while for $T > \epsilon_F^*(\delta)$, the system should become an incoherent metal, i.e., a Mott insulator with thermal excitations in the gap. However, as we shall see, the crossover in the intermediate T regime investigated here occurs rather differently from this naive expectation.

In the left panel of Fig.2 we plot the evolution of the DOS for a series of very low dopings. The bottom most curve shows that sufficient doping results in a heavy Fermi liquid metal with a narrow quasiparticle centered at the Fermi energy. At the other end, the top most

curve shows a virtually insulating Mott state with negligible DOS at the Fermi energy and a large charge gap ($\sim U$). The doping of this state is extremely small ($\approx 0.3\%$) and the high temperature does not allow for a narrow coherent quasiparticle band. The interesting aspect of the crossover between these states is the anomalous evolution of the quasiparticle peak, shown in detail in the top right panel of Fig.2. There are two features worth noticing: in contrast to the scenario described earlier the quasiparticle peak persists down to very low values of doping $\delta \sim 0.01$, in a regime where $T \gg \delta D$. The second unexpected feature is that the position of quasiparticle peak shifts away from the Fermi energy. The asymmetry that the peak develops around $\omega = 0$ can be physically understood as due to the relative suppression of particle propagation with respect to hole propagation in a lightly hole doped Mott insulator (or viceversa). This anomalous behavior of the quasiparticle peak leads us to term the state in the crossover regime as a “bad metal” (BM). Interestingly, the asymmetry around $\omega = 0$ was already observed in the low frequency features of the AF state. While its origin in the AF state might have not seemed obvious, it appears clearer under the new light shed by the results in the PM state. It is perhaps worth mentioning that the anomalous shift of the quasiparticle peak off the Fermi energy is reminiscent of the phenomenology of the pseudo-gap state observed in photoemission studies of underdoped cuprates [7]. Our results for the evolution of the DOS are summarized in the phase diagram of the Fig.2.

Finally, we consider the critical behavior of the occupation number $n = 1 - \delta$ and the staggered magnetization m at the AF/PM boundary. The QMC results are shown in Fig.3. The most striking feature is that the slope $\partial n / \partial \mu$ which determines the electronic *charge* compressibility, shows a dramatic enhancement on the AF side of the transition line which corresponds to the AF metal state that we described before. More significantly, the enhancement rapidly increases as T is lowered. For instance, reducing T by 50% yields a 7 fold increase in the compressibility. Our results are not inconsistent with a divergence at a low finite temperature, but due to strong numerical fluctuations at the transition boundary obtaining reliable data at lower T becomes impractical. Such a divergence might be a signal of an instability to phase separation. In any case, this striking behavior is expected to have clear experimental consequences promoting phase separation (PS), anomalies in sound velocity propagation [19] and structural or electronic instabilities [20] among others. Remarkably, while the compressibility shows this anomalous behavior, the staggered magnetization in contrast displays the expected critical behavior at the transition. We find $m \propto (\mu - \mu_c)^{1/2}$

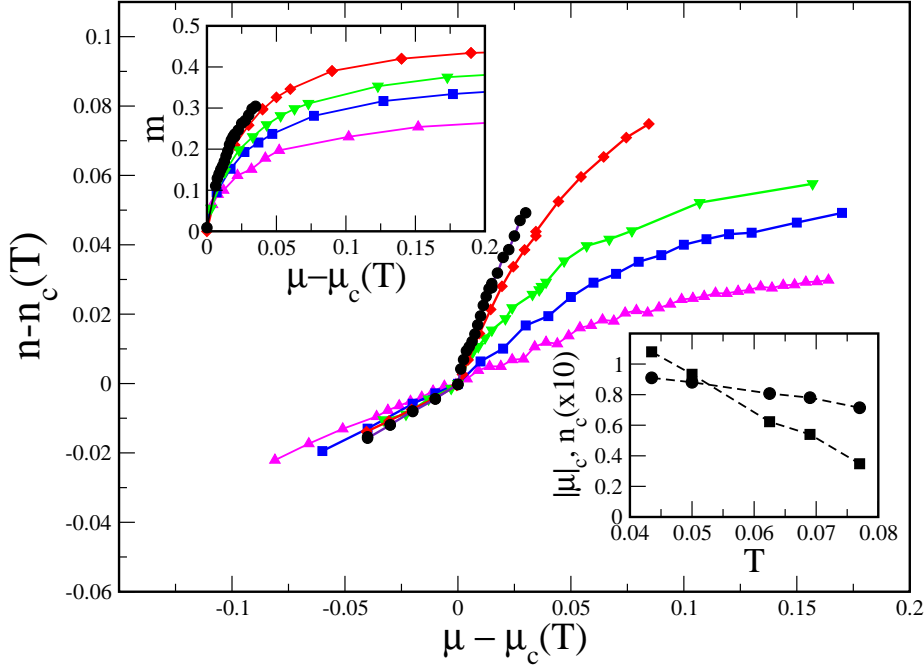


FIG. 3: Occupation $n - n_c(T)$ as a function of the shifted chemical potential $\mu - \mu_c(T)$ from QMC at $U = 3.125$ and different T values. $T = 0.043$ (black circles), 0.05 (red diamonds), 0.0625 (green down triangles), 0.069 (blue squares) and 0.077 (purple up triangles). Error bars are about the size of the symbols. Top inset: m vs. $\mu - \mu_c(T)$. Bottom inset: $|\mu_c|$ (circles) and n_c (squares) as a function of T .

for all T .

In conclusion, we study the behavior of a doped Mott-Hubbard insulator within the framework of DMFT. Various interesting regimes were found as the model is doped away from both the half-filled antiferromagnetic and paramagnetic states. In particular, the study of the DOS allowed us to identify a strongly correlated AF metal state and a PM “bad metal” one. The crossover through a “bad metal” state was identified. Along the AF/PM critical line a compressibility anomaly was observed with a dramatic enhancement at lower temperatures. The relevance of our results for the interpretation of various experiments was pointed out. Though a calculation of the optical conductivity in these regimes would be clearly interesting, it is technically challenging and is beyond the scope of the present paper. Interesting perspectives are opened by this work such as the investigation of superconducting

instabilities and similar features in more realistic multiorbital models.

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